

No new matter has been added by these amendments; therefore, Applicants respectfully request that examination continue on the claims as amended herewith.

**Rejection under 35 U.S.C. § 103**

The Office Action rejected claims 1-6 under 35 U.S.C. § 103(a) as allegedly being unpatentable over Halloran (U.S. Patent 6,071,975). Specifically, the Office Action alleged that Halloran teaches oil-in-water emulsions in which the continuous phase can contain diols in an amount within the claimed range. Applicants respectfully traverse this rejection to the extent that it is applied to the amended claims.

Applicants first note that Halloran does not teach or suggest silicone polymer emulsions. Instead, Halloran only discloses the preparation of microemulsions, which are different from emulsions. As is shown from Duncan J. Shaw, "Introduction to Colloid and Surface Chemistry," Fourth Edition, Butterworth-Heinemann Ltd., Oxford, 1992, pp. 262-263 and 89-90, a copy of which is enclosed as Exhibit A, an emulsion is defined as a dispersed system where the globules of the dispersed phase have a size range of from 0.1 to 10  $\mu\text{m}$ , *i.e.*, 100 to 10,000 nm, and a microemulsion has a dispersed phase with droplets in the 0.01 to 0.1  $\mu\text{m}$  size range, *i.e.*, 10 to 100 nm. This definition is confirmed by Halloran, which states that "an emulsion is considered as containing structures as having an average diameter of more than 100 nm [0.1  $\mu\text{m}$  ], whereas a microemulsion contains structures having an average diameter of less than 100 nm...most preferably less than 10 nm." (Halloran at col. 2, lines 15-26.) Accordingly, contrary to the statements in the Office Action, Halloran does not teach "a silicone oil in water emulsion in which the continuous phase, the water phase, can contain diols in an amount within the claimed phase." (See Office Action at page 2.)

Further, Halloran teaches away from an emulsion, as recited in the claims, because this reference is directed towards optically clear compositions. (See Halloran at col. 1, lines 17-21.) As noted by Halloran, the clarity of a composition is controlled by the particle size of

the dispersed phase because the scattering of light is dependent on the particle size. (Halloran at col. 2, lines 27-33.) This phenomenon is also discussed in the Exhibit A, which states that "the visual appearance of an emulsion reflects the influence of droplet size on light scattering, and varies from milky-white-opaque, with large droplets, through blue-white, then gray-translucent, to transparent, with small microemulsion droplets." In other words, since the preparation of emulsions would be contrary to Halloran's objectives of obtaining optically clear compositions, Halloran teaches away from an emulsion, as recited in the claims.

Moreover, Halloran states that emulsions are inherently unstable, but that microemulsions are stable indefinitely. (Halloran at col. 2, lines 33-36.) Halloran also states that clear microemulsions exhibit a variety of advantages and beneficial properties such as clarity, very small structure size, ultra-low interfacial tensions, etc. (Halloran at col. 11, lines 4-11.) Therefore, in addition to the discussion above, Halloran expressly discourages the skilled artisan from preparing emulsions for at least this additional reason.

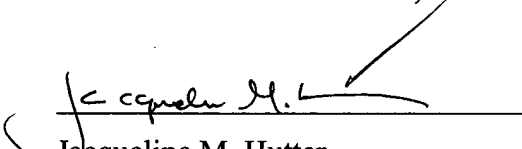
Additionally, Halloran only discloses oil-in-water microemulsions in which the continuous phase of the microemulsion may contain 50 wt. % water, 5 wt. % surfactant, and 15 wt. % of a co-surfactant that can be a diol. (See Halloran at col. 4, lines 50-59 and col. 6, lines 37-41.) Halloran does not disclose using more than 15 wt. % of the co-surfactant. However, the Office Action makes an unsupported allegation that Halloran's disclosed 15 wt. % of co-surfactant suggests the "about 25 % by weight" recited in the claims. Thus, even assuming that Halloran suggested the preparation of emulsions (which it does not), since the Office Action does not provide any support for the contention that Halloran's 15 % by weight co-surfactant would suggest or motivate the "about 25 % by weight" recited by Applicants, the Office Action does not set forth a *prima facie* case of obviousness against the claimed invention.

CONCLUSION

Pursuant to the above Amendments and Remarks, reconsideration and allowance of the pending application is believed to be warranted.

A Credit Card Payment Form PTO-2038 authorizing payment in the amount of \$750.00, for the fee under 37 C.F.R. § 1.17(e) and a Request for Continued Examination are enclosed. This amount is believed to be correct; however, the Commissioner is hereby authorized to charge any additional fees that may be required, or credit any overpayment to Deposit Account No. 14-0629.

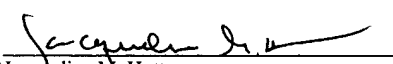
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I hereby certify that this AMENDMENT AND RESPONSE ACCOMPANYING RCE is being deposited with the United States Postal Service as first class mail in an envelope addressed to: BOX RCE, Commissioner for Patents, Washington D.C. 20231 on the date shown below.

  
Jacqueline M. Hutter

3/6/03  
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TC 1700

ATTORNEY DOCKET NO.: 05015.0209U3  
APPLICATION NO.: 09/997,455

VERSION WITH MARKINGS TO SHOW CHANGES MADE

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1. (Twice Amended) A silicone polymer emulsion consisting essentially of:
  - a. [a] silicone polymer particles;
  - b. a surfactant; and
  - c. a liquid continuous phase comprising a diol, wherein the diol comprises from about 25 to about 100% by weight of the continuous phase, [and] wherein the silicone polymer particles are [is] dispersed in the continuous phase, and wherein the silicone polymer particles have a size range of from about 0.1 to 10 microns.
  
5. (Twice Amended) The silicone polymer emulsion of claim 1, wherein the surfactant comprises alkali metal[,] or ammonium salts of alkyl, aryl or alkylaryl sulfonates, sulfates, phosphates, alkyl and alkylaryl polydiol ethers, ethoxylation products of lauryl, oleyl and stearyl alcohols, alkyl phenol glycol ethers, ethoxylation products of octyl or nonylphenol, or a mixture thereof.

To Ann

# Introduction to Colloid and Surface Chemistry

Fourth edition

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# 10 Emulsions and foams

## Oil-in-water and water-in-oil emulsions<sup>123-126</sup>

An emulsion is a dispersed system in which the phases are immiscible or partially miscible liquids. The globules of the dispersed liquid in the usual type of emulsion (sometimes now called a macroemulsion) are usually between 0.1  $\mu\text{m}$  and 10  $\mu\text{m}$  in diameter, and so tend to be larger than the particles found in sols.

The practical application of emulsions and emulsion technology is considerable, and includes foodstuffs (especially dairy produce), pharmaceutical preparations, cosmetics, agricultural sprays and bituminous products. Emulsions enable the dilution of an expensive or concentrated ingredient with an inexpensive, but immiscible, diluent. For example, water-insoluble agrochemicals are generally marketed in the form of oil in water emulsions which can be diluted with water by the user in order to permit even, low-level application. Emulsions also allow enhanced control over other factors, such as rheology and the various manifestations of chemical reactivity (including taste, odour and toxicity). A large volume of technological information on emulsions exists, much of it in private files. In general, empirical knowledge is well ahead of fundamental understanding.

Microemulsions, with droplet diameters of 0.01–0.1  $\mu\text{m}$ , can also be prepared and these are currently the subject of much fundamental investigation and new applications (e.g. oil recovery from porous rocks). Whether microemulsions should be regarded as true emulsions or as swollen micelles (see section on solubilisation, page 89) is a matter of controversy.

The visual appearance of an emulsion reflects the influence of droplet size on light scattering, and varies from milky-white-opaque,

with large droplets, through blue-white, then gray-translucent, to transparent, with small microemulsion droplets.

In nearly all emulsions, one of the phases is aqueous and the other is (in the widest sense of the term) an oil. If the oil is the dispersed phase, the emulsion is termed an *oil-in-water* (O/W) emulsion; if the aqueous medium is the dispersed phase, the emulsion is termed a *water-in-oil* (W/O) emulsion. There are several methods by which the emulsion type may be identified.

1. In general, an O/W emulsion has a creamy texture and a W/O emulsion feels greasy.
2. The emulsion mixes readily with a liquid which is miscible with its dispersion medium.
3. The emulsion is readily coloured by dyes which are soluble in the dispersion medium.
4. O/W emulsions usually have a much higher electrical conductivity than W/O emulsions.

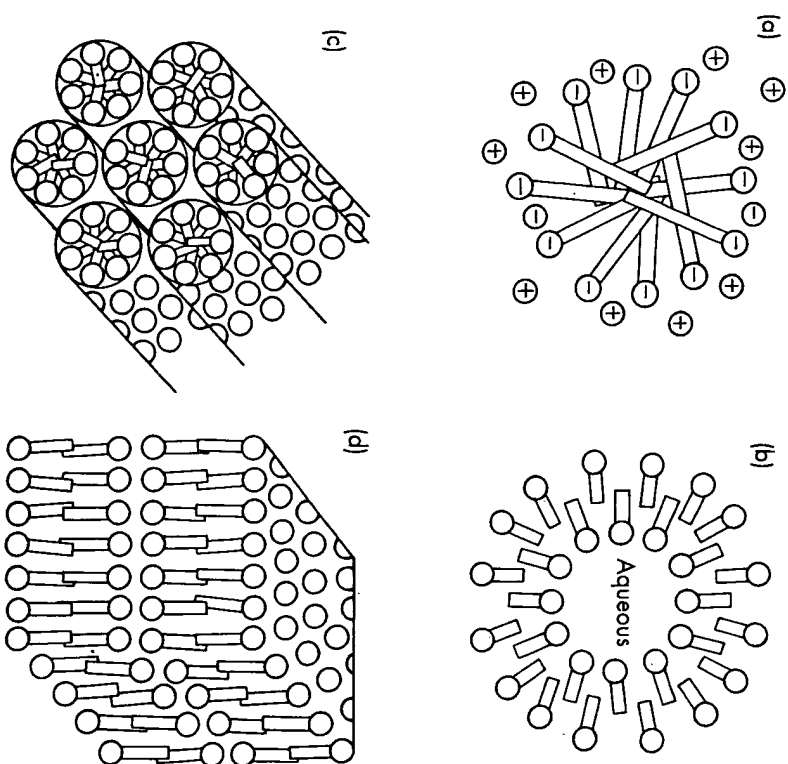
### Emulsifying agents and emulsion stability

Probably the most important physical property of an emulsion is its stability. The term 'emulsion stability' can be used with reference to three essentially different phenomena – creaming (or sedimentation), coagulation and a breaking of the emulsion due to droplet coalescence. Creaming results from a density difference between the two phases and is not necessarily accompanied by droplet coagulation, although it facilitates this process.

Droplet collisions may result in coagulation, which, in turn, may lead to coalescence into larger globules. Eventually, the dispersed phase may become a continuous phase, separated from the dispersion medium by a single interface. The time taken for such phase separation may be anything from seconds to years, depending on the emulsion formulation and manufacturing conditions.

Assessment of the stability of an emulsion against coalescence involves droplet counting<sup>218</sup>. The most unequivocal method (but one which is rather laborious) is to introduce a suitably diluted sample of the emulsion into a haemocytometer cell and count the microscopically visible particles manually.

The Coulter counter affords a convenient indirect technique for



**Figure 4.14** Micellar structures. (a) Spherical (anionic) micelle. This is the usual shape at surfactant concentrations below about 40 per cent. (b) Spherical vesicle bilayer structure (see also Figure 4.28), which is representative of the living cell. (c) and (d) Hexagonal and lamellar phases formed from cylindrical and lamellar micelles, respectively. These, and other structures, exist in highly concentrated surfactant solutions

micelle,  $m$ , and the number of carbon atoms per hydrocarbon chain,  $n$ , are approximately related as follows:

$n$	12	14	16	18
$m$	33	46	60	78

Lamellar and cylindrical models, in contrast, provide no satisfactory mechanism by which the size of the micelles might be limited.

3. For diffusion reasons, solubilisation (see next section) would not take place readily if the micelle were solid.

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As mentioned above, the length of the surfactant's hydrocarbon chain will dictate the radius of a spherical micelle. This in turn determines the spacing of the outer polar groups. On this basis, for example, a dodecyl sulphate micelle surface would be expected to be approximately one-third sulphate groups and two-thirds hydrocarbon. The results of neutron scattering studies are consistent with this expectation. In an ionic micelle, the tendency of this hydrocarbon-water interfacial area to contract is balanced by head-group repulsion. Addition of electrolyte reduces this head-group repulsion, thus favouring an area per head-group that is smaller than the geometric optimum for a spherical micelle. Under such conditions, the micelle is likely to distort to a non-spherical shape.

There is evidence from nuclear magnetic resonance spectroscopy and partial molar volume measurements<sup>153-154</sup> which points to the possible existence of bound water in the micelle interior in the region of the first few  $\text{CH}_2$  groups in from the polar head groups. The hydrocarbon interior of the micelle may, therefore, be considered in terms of an outer region which may be penetrated by water and an inner region from which water is excluded.

### Solubilisation<sup>51</sup>

Surfactant solutions above the c.m.c. can solubilise otherwise insoluble organic material by incorporating it into the interior of the micelles; for example, the dye xlenol orange dissolves only sparingly in pure water but gives a deep red solution with sodium dodecyl sulphate present above its c.m.c.

The balance of electrostatic and hydrophobic interactions can be such as to cause the locus of solubilisation to be anywhere in the micelle from close to the surface to the inner core.

Solubilisation is of practical importance in the formulation of pharmaceutical and other products containing water-insoluble ingredients<sup>51</sup>, detergency, where it plays a major role in the removal of oily soil (pages 166-176), emulsion polymerisation (page 17) and micellar catalysis of organic reactions<sup>52</sup>.

In micellar catalysis, reactant must be solubilised at a location near to the micelle surface where it is accessible to reagent in the aqueous

phase. The strong electrostatic interactions which are likely at this location may influence the nature of the transition state and/or reactant concentration; for example, cationic micelles may catalyse reaction between a nucleophilic anion and a neutral solubilised substrate.

### Surface behaviour

Figure 4.13 illustrates how a highly surface-active material such as sodium dodecyl sulphate lowers the surface tension of water quite appreciably even at low concentrations. The discontinuity in the  $\gamma$ -composition curve is identified with the c.m.c., beyond which there is an additional mechanism for keeping hydrocarbon chains away from water surfaces – i.e. by locating them in the interior of the micelles. Since the micelles themselves are not surface-active, the surface tension remains approximately constant beyond the c.m.c. The minimum in the  $\gamma$ -composition curve, shown by the dashed curve, is typical of measurements which have been made on surfactant solutions and in apparent violation of the Gibbs equation, since it suggests desorption over the small concentration range where  $d\gamma/dc$  is positive. This anomaly is attributed to traces of impurity such as dodecanol, which is surface-adsorbed below the c.m.c. but solubilised by the micelles beyond the c.m.c. With sufficient purification the minimum in the  $\gamma$ -composition curve can be removed. Beyond the c.m.c., where  $d\gamma/dc \sim 0$ , application of the Gibbs equation might suggest almost zero adsorption; however,  $d\gamma/da$ , where  $a$  represents the activity of single surfactant species, is still appreciably negative,  $a$  changing little above the c.m.c.

### Conductance

Micelle formation affects the conductance of ionic surfactant solutions for the following reasons:

1. The total viscous drag on the surfactant molecules is reduced on aggregation.
2. Counter-ions become kinetically a part of the micelle, owing to its high surface charge (see Chapter 7), thus reducing the number of counter-ions available for carrying the current and also lowering the net charge of the micelles. Typically, 50 to 70% of the counter-

ions are held in the Stern layer; even so, the zeta potential of an ionic micelle is usually high.

3. The retarding influence of the ionic atmospheres of unattached counter-ions on the migration of the surfactant ions is greatly increased on aggregation.

The last two factors, which cause the molar conductivity to decrease with concentration beyond the c.m.c., normally outweigh the first factor, which has the reverse effect (see Figure 4.13). When conductance measurements are made at very high field strengths the ionic atmospheres cannot re-form quickly enough (Wien effect) and some of the bound counter-ions are set free. It is interesting to note that under these conditions the molar conductivity increases with concentration beyond the c.m.c.

### Sharpness of critical micelle concentrations

There are two current theories relating to the abruptness with which micellisation takes place above a certain critical concentration<sup>53,155</sup>.

The first of these theories applies the law of mass action to the equilibrium between unassociated molecules or ions and micelles, as illustrated by the following simplified calculation for the micellisation of non-ionic surfactants. If  $c$  is the stoichiometric concentration of the solution,  $x$  is the fraction of monomer units aggregated and  $m$  is the number of monomer units per micelle,

$$mX = (X)_m \\ c(1-x) \quad cx/m$$

Therefore, applying the law of mass action,

$$K = \frac{cx/m}{[c(1-x)]^m} \quad (4.23)$$

For moderately large values of  $m$ , this expression requires that  $x$  should remain very small up to a certain value of  $c$  and increase rapidly thereafter. The sharpness of the discontinuity will depend on the value of  $m$  ( $m = \infty$  gives a perfect discontinuity). If this treatment is modified to account for the counter-ions associated with an ionic micelle, then an even more abrupt discontinuity than the above is predicted.